

# Evidence of electro-active excitation of the spin cycloid in TbMnO<sub>3</sub>

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Terahertz electromagnetic excitations in the multiferroic TbMnO<sub>3</sub> at the field-induced magnetic transition are investigated for different orientations of the magnetic cycloid. In addition to the electromagnon along the *a*-axis, the detailed polarization analysis of the experimental spectra suggests the existence of an electro-active excitation for ac electric fields along the crystallographic *c*-axis. This excitation is possibly the electro-active eigenmode of the spin cycloid in TbMnO<sub>3</sub>, which has been predicted within the inverse Dzyaloshinskii-Moriya mechanism of magnetoelectric coupling.

Multiferroics represent an intriguing class of materials in which magnetism and electricity are strongly coupled. This magnetoelectric coupling leads to a mutual influence of magnetic and electric ordering, which results in a rich new physics and in various possible applications [1–4]. Recently, a new class of multiferroics has attracted enormous interest because the ferroelectric polarization in these systems is induced by a cycloidal ordering of the magnetic moments [5–7]. The magnetic and electric order in these materials are accompanied by coupled spin-lattice vibrations [8] showing strong electric dipole activity and termed electromagnons [9, 10]. Initially detected in GdMnO<sub>3</sub> and TbMnO<sub>3</sub> [11], the electromagnons appear to exist in cycloidal magnetic phases of many different multiferroics [12–19]. In most cases, distinct structure of these excitations is seen including two or more separate modes. For example, in TbMnO<sub>3</sub> one can observe at least 3 excitations: two with the energy around  $\simeq 2$  meV ( $10\text{--}20\text{ cm}^{-1}$ ) and the third one at  $\simeq 8$  meV ( $60\text{ cm}^{-1}$ ).

Ferroelectric polarization in spiral magnets has been successfully explained on the basis of the inverse Dzyaloshinskii-Moriya (DM) coupling between the magnetic moments [20–22]. Within this model a spiral spin structure lowers the symmetry of the system leading to an effective magnetoelectric coupling term and an electrical polarization *P* in the form

$$P \sim e_{ij} \times S_i \times S_j \quad . \quad (1)$$

Here *S<sub>i</sub>* and *S<sub>j</sub>* are the neighboring spins and *e<sub>ij</sub>* is the lattice vector connecting them. Consequently, it was reasonable to assume the same mechanism to explain the existence of the electromagnons [23]. In this model the electromagnons are the eigenmodes of the spin cycloid which can be excited by electric component of the electromagnetic wave [9]. However, after initial success of this explanation a number of experimental results contradicted the predictions of the model. The basic example can be given by TbMnO<sub>3</sub> at low temperatures: the DM model predicts that the excitation conditions for the electromagnon should be tied to the magnetic cycloid, i.e. for the bc-plane oriented cycloid the electric polariza-

tion must be parallel to the *c*-axis (*P*||*c*) and the electromagnons should be excited for ac electric fields parallel to the *a*-axis ( $\tilde{e}$ ||*a*). Accordingly, for the *ab*-plane oriented cycloid the polarization *P*||*a* and electromagnons for  $\tilde{e}$ ||*c* should be observed. This basically corresponds to the interchange of the *a* and *c*-axes in the excitation conditions. Although the orientation of the static electric polarization has been confirmed in many experiments [5, 24, 25], the excitation conditions for the electromagnon contradicted the predictions of the model: the selection rules for the excitations remained the same (i.e.  $\tilde{e}$ ||*a*) independently of the orientation of the cycloid [11–19]. In order to account for this contradiction, a model based on Heisenberg exchange coupling between spins has been proposed recently [18, 26, 27]. According to this model, the structural peculiarities of orthorhombic multiferroic manganites lead to the selection rules  $\tilde{e}$ ||*a* regardless of the orientation of the spin cycloid.

The Heisenberg exchange model assigns the high energy electromagnons observed between  $60\text{ cm}^{-1}$  and  $80\text{ cm}^{-1}$  to the zone edge magnons [18, 26–28], but the nature of the lower energy electromagnons close to  $20\text{ cm}^{-1}$  remains uncertain. It seems reasonable to recall the statements of the DM model and to assume that the low-frequency electromagnons are somehow related to the excitations of the spin cycloid observed by inelastic neutron scattering (INS) [29–31] because the characteristic frequencies coincide closely [29, 32]. This assumption is supported by a recent observation of the magnetic excitation channel for electromagnons [32] which can be also seen as antiferromagnetic resonances (AFMR) within certain excitation conditions. However, in order to prove this assumption, the predicted excitation conditions for the eigenmodes of the cycloid should be found experimentally. Most specifically, one should expect the excitation conditions along the *c*-axis if the spin cycloid is oriented in the *ab*-plane. Such excitation conditions were not observed up to now [26, 32]. A possible reason for this fact is the weakness of the dielectric contribution of the spin modes within the DM mechanism. In order to resolve this experimental difficulty, TbMnO<sub>3</sub> seems to be an ideal candidate, because the magnetic cycloid can be

rotated between  $ab$ -plane and  $bc$ -plane in external magnetic fields. Thus the ultimate experiment must follow the  $c$ -axis response at the magnetically induced rotation of the cycloid.

In this work we have carried out detailed polarization analysis of terahertz excitations in  $\text{TbMnO}_3$ . Special attention has been paid to the selection rules of different excitations upon the rotation of the spin cycloid from  $bc$ - to  $ab$ -plane in external magnetic field  $B \parallel b$ -axis. It was found that a new excitation arises in the high-field phase with excitation conditions  $\tilde{e} \parallel c$  and  $\tilde{h} \parallel a$ . We argue that this excitation could not be explained by purely magnetic contribution but carries substantial electric component.

The transmittance experiments at terahertz frequencies ( $3 \text{ cm}^{-1} < \nu < 30 \text{ cm}^{-1}$ ) have been carried out in a Mach-Zehnder interferometer arrangement [33, 34] which allows measurements of amplitude and phase shift in a geometry with controlled polarization of radiation. The experiments in external magnetic fields up to 8 T have been performed in a superconducting split-coil magnet with polypropylene windows. Single crystals of  $\text{TbMnO}_3$  have been grown using the floating-zone method with radiation heating. The samples were characterized using X-ray, magnetic, dielectric and optical measurements [9, 35]. The results of these experiments including the magnetic phase diagram are closely similar to the published results [24].

The paramagnetic phase in  $\text{TbMnO}_3$  above 40 K is followed by a sinusoidally-modulated antiferromagnetic state which transforms below 28 K into the cycloidal spin structure oriented within the  $bc$ -plane [36, 37] (inset in Fig. 1). According to the symmetry analysis [6, 22] and Eq. (1), static electric polarization along the  $c$ -axis is allowed in the low-temperature phase. In external magnetic fields the spin-cycloid rotates from  $bc$ -plane towards the  $ab$ -plane [25, 31]. Correspondingly, the electric polarization rotates from  $P \parallel c$ -axis to the  $P \parallel a$ -axis [5, 24, 25].

Dynamic experiments on  $\text{TbMnO}_3$  in external magnetic fields reveal that the  $c$ -axis properties are indeed sensitive to the orientation of the cycloid. The examples of such changes are shown in Fig. 1 which represents the terahertz properties of  $\text{TbMnO}_3$  in external magnetic field  $B \parallel b$  inducing the transition from  $bc$ -plane to  $ab$ -plane oriented cycloid. The magnetic field scans in these experiments were made in the geometry with  $\tilde{e} \parallel c$  and  $\tilde{h} \parallel a$  and at  $T = 10 \text{ K}$ . The data are represented as refractive index  $n + i\kappa = \sqrt{\epsilon\mu}$  as both electric and magnetic contributions could be mixed in this experimental geometry. The transition from  $bc$ -plane to  $ab$ -plane cycloid in the high magnetic fields is clearly seen. Already at this point it is evident that the observed changes are strongly frequency dependent. Here the data at  $4.7 \text{ cm}^{-1}$  is influenced by a Tb-mode around  $5 \text{ cm}^{-1}$  [32] which disappears in the high-field phase with the  $ab$ -plane cycloid. This leads to a substantial decrease of the absorption ( $\kappa(4.7 \text{ cm}^{-1})$ ) and reveals a bit complicated structure in

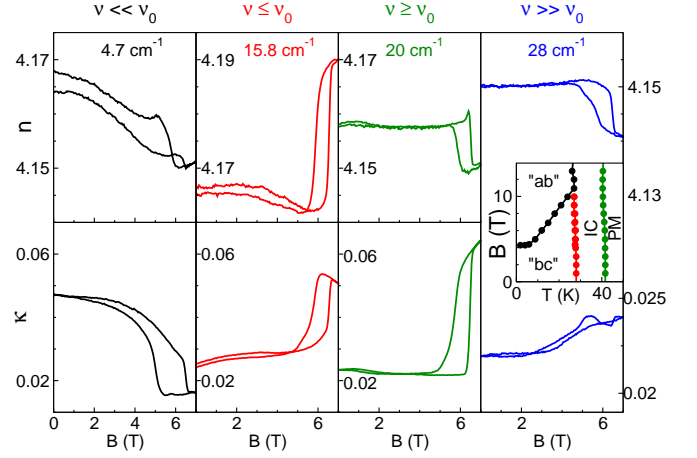


FIG. 1: Dependence of refractive index ( $n = \text{Re}(\sqrt{\epsilon\mu})$ , upper panels) and absorption coefficient ( $\kappa = \text{Im}(\sqrt{\epsilon\mu})$ , lower panels) in  $\text{TbMnO}_3$  on magnetic field  $B \parallel b$ -axis at various frequencies on crossing the phase transition from the  $bc$ - to the  $ab$ -oriented magnetic cycloid. Polarization of incident wave is  $\tilde{e} \parallel c$ ,  $\tilde{h} \parallel a$ , where  $\tilde{e}$  and  $\tilde{h}$  are electric and magnetic ac fields of the electromagnetic wave. The inset shows  $B$ - $T$  phase diagram of  $\text{TbMnO}_3$  for  $B \parallel b$  [24]. “ $ab$ ” and “ $bc$ ” denote  $ab$ -plane and  $bc$ -plane oriented cycloids, respectively, PM - paramagnetic and IC - sinusoidal phases.

refractive index below  $10 \text{ cm}^{-1}$ . The changes observed at  $4.7 \text{ cm}^{-1}$  can be well understood assuming a suppression of a Lorentzian mode situated between  $5$  and  $6 \text{ cm}^{-1}$ .

Three higher frequency scans ( $15.8$ ,  $20$ , and  $28 \text{ cm}^{-1}$ ) in Fig. 1 show more systematics, and can be reduced to a growth of the absorption mode around  $20 \text{ cm}^{-1}$  in the phase with the  $ab$ -plane cycloid. This is a typical behavior for a Lorentz oscillator which appears close to  $20 \text{ cm}^{-1}$  simultaneously with the  $ab$ -plane cycloid. Indeed, strong additional absorption arises near the frequency  $\simeq 20 \text{ cm}^{-1}$  and is substantially reduced above and below this frequency (lower panels,  $28 \text{ cm}^{-1}$  and  $16 \text{ cm}^{-1}$ , respectively). In addition to a strongly increased absorption around the resonant frequency, the refractive index in Lorentzian model may in rough approximation be simplified to the following statements:  $n(\nu) \approx n_\infty + \Delta n$  for  $\nu < \nu_0$  and  $n(\nu) \approx n_\infty - \Delta n(\nu_0/\nu)^2$  for  $\nu > \nu_0$ . Here  $n_\infty$  is the contribution from the high-frequency processes,  $\Delta n$  reflects the strength of the Lorentzian mode, and  $\nu_0 \approx 20 \text{ cm}^{-1}$  is the resonance frequency. Therefore, after the growth of the Lorentzian ( $\Delta n = 0 \rightarrow \Delta n \neq 0$ ) at the magnetic transition we expect an increase of the refractive index below the resonant frequency and a decrease above it. This is roughly the behavior of the refractive index, observed at  $15.8 \text{ cm}^{-1}$  ( $\nu < \nu_0$ ) and  $28 \text{ cm}^{-1}$  ( $\nu > \nu_0$ ). The main changes, seen in Fig. 1, appears at the magnetic transition and are therefore related to the rotation of the magnetic cycloid. However, better understanding of the underlying processes can be obtained within a direct

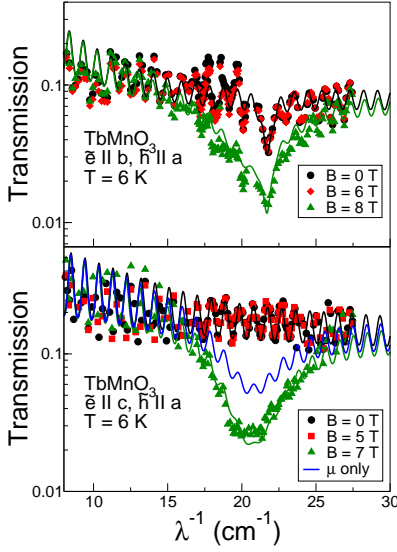


FIG. 2: Transmittance spectra of TbMnO<sub>3</sub> in external magnetic fields  $B||b$  for different experimental geometries. Symbols are experimental data and solid lines are fits with Lorentz oscillators as discussed in the text.

analysis of the spectra in the relevant frequency range as presented below.

Figure 2 shows the field dependent spectra for two different geometries of the experiment. The thicknesses of the samples are similar for both orientations: 1.24 mm (upper panel) and 1.33 mm (lower panel), respectively. The spectra in the lower panel with  $\tilde{e}||c$  and  $\tilde{h}||a$  correspond well to the known results [26, 32] and show a mode at about 21 cm<sup>-1</sup> which appears after the field induced reorientation of spin cycloid to the  $ab$ -plane. Based on the weakness of this mode, both in Ref. [26] and in Ref. [32] it has been concluded that the mode around 21 cm<sup>-1</sup> is of purely magnetic origin and represent an antiferromagnetic resonance of the magnetic cycloid for  $\tilde{h}||a$ -axis. Indeed, the strength of this mode ( $\Delta\epsilon \sim 0.05$ , assuming electric origin) is extremely weak compared to electromagnon observed for  $\tilde{e}||a$  ( $\Delta\epsilon \sim 2$ ) [9, 11]. The mode in Fig. 2 is observed for the  $ab$ -plane cycloid and within  $\tilde{h}||a$  excitation conditions. Tracing this mode back into the  $bc$ -oriented cycloid in zero external fields, it can be expected to originate from the excitation conditions  $\tilde{h}||c$ . (This corresponds to the interchanging of the  $a$ -axis and  $c$ -axis). Indeed, an AFMR mode excited for  $\tilde{h}||c$  of the similar strength has been observed around 21 cm<sup>-1</sup> in the phase with the  $bc$ -plane cycloid [32].

A careful comparison of both panels in Fig. 2 reveals interesting difference between two excitation conditions. The strength of the mode in the geometry where  $\tilde{e}||b$  is roughly the half of that where  $\tilde{e}||c$ . This strongly suggests that for geometry in which  $\tilde{e}||c$  the electric dipole contribution is indeed measurable and represent the previously unobserved  $\tilde{e}||c$  counterpart of the electromagnon. These

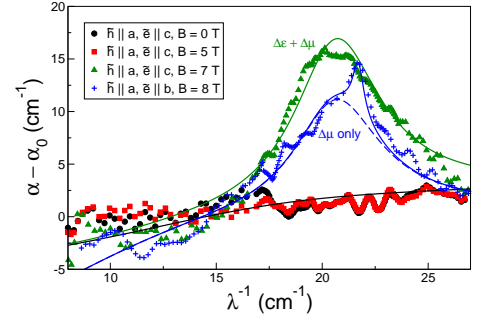


FIG. 3: Linear absorption coefficient  $\alpha = 4\pi\kappa\lambda^{-1}$  in TbMnO<sub>3</sub> for different polarizations showing the emergence of an additional mode in the phase with the  $ab$ -cycloid (green triangles and blue crosses). Symbols are experimental data calculated from transmittance spectra as described in the text. Lines are model calculations using Lorentz oscillators with the same parameters as in Fig. 2. The data are shifted vertically to eliminate background absorption in different samples.

results agree well with the original explanation of the electromagnons as electrically active eigenmodes of the cycloidal structure [9, 23].

In order to make the discussion quantitative, the experimental spectra in the upper panel of Fig. 2 were fitted with magnetic Lorentz oscillator. If we now take the parameters of the mode from the geometry with  $\tilde{e}||b$  and plot the expected transmittance spectra for the geometry  $\tilde{e}||c$  we obtain the absorption value which is too weak compared to the experiment (the “ $\mu$  only” curve in the lower panel of Fig. 2). The only possible explanation is that this mode has distinct non-zero electric contribution along the  $c$ -axis. The actual fit for this geometry was obtained by taking parameters of the magnetic oscillator from  $\tilde{e}||b$ ,  $\tilde{h}||a$  geometry and adding an electric oscillator with the same resonance frequency  $\nu_0 = 20.7$  cm<sup>-1</sup> and line width  $\gamma = 4.9$  cm<sup>-1</sup> as the magnetic one. The reasoning behind this assumption is that both contributions are electric and magnetic parts of the same eigenmode of the spin cycloid. The strengths of both components is given by  $\Delta\mu = 0.0038$  and  $\Delta\epsilon = 0.05$ , respectively.

Figure 3 shows the linear absorption coefficients  $\alpha = 4\pi\kappa\lambda^{-1}$  which allow direct comparison of different experimental geometries. Symbols were calculated from transmittance spectra  $T$  using the expression  $T = (1 - R)^2 \exp(-\alpha d)$ , where  $R = |(\sqrt{\epsilon\mu} - 1)/(\sqrt{\epsilon\mu} + 1)|^2$  is the reflectance on the boundary between the air and the sample, and  $d$  is the sample thickness. Solid lines are model calculations using the same parameters as in Fig. 2. One can see again that the mode at 21 cm<sup>-1</sup> is stronger in the case of  $\tilde{e}||c$  and  $\tilde{h}||a$  excitation. (A weak narrow mode seen close to 22 cm<sup>-1</sup> for  $\tilde{e}||b$ ,  $\tilde{h}||a$  geometry is possibly due to impurities in the sample. The strength of this mode is at least an order of magnitude smaller than the strength of the broad mode and doesn’t change the overall picture.)

The mode intensity for the “main”  $\tilde{e}||a$  electromagnon ( $\Delta\epsilon_a \simeq 2$  [11]) is about 40 times stronger than electric contribution along the  $c$ -axis ( $\Delta\epsilon_c \simeq 0.05$ ) observed in the present experiment. The large electromagnon absorption along the  $a$ -axis was one of the challenging questions in explaining its origin. The relatively weak static electric polarization doesn’t fit well with the large dielectric absorption of electromagnon if both are caused by Dzyaloshinskii-Moriya interaction [26]. On the contrary, the Heisenberg exchange mechanism [18, 26, 27] seems to explain well the intensities of at least the high-frequency electromagnons above  $50 \text{ cm}^{-1}$ . In this model the edge-zone magnon couples to alternating orthorhombic distortions at oxygen sites via symmetric Heisenberg exchange interaction. This leads to the coupling of the zone edge magnon to homogeneous electric fields along the  $a$ -axis. As the symmetric interaction is much stronger than the relativistic DM coupling, the hybridized electromagnon has enough strength to explain the experimental intensities for  $\tilde{e}||a$ . Much weaker [26] DM component cannot be seen in this experimental geometry because of the dominance of the intensity induced by the Heisenberg exchange coupling. On the contrary, rotating the magnetic cycloid towards the  $ab$ -plane, both contributions can be well separated experimentally. The Heisenberg exchange part remain oriented along the  $a$ -axis, as confirmed by different experimental groups [15, 19, 26, 32]. The weak DM electromagnon rotates with the cycloid and can be clearly observed in the present experiment as electric contribution along the  $c$ -axis.

One remaining question is: why we observe only one mode in the high-field phase? The probable reason is that one of two modes is too weak and is not seen in the spectra. This argument is supported by recent inelastic neutron scattering experiments [31]. In these experiments the modes of the  $ab$ -plane spin cycloid have been investigated. Although this  $ab$ -plane orientation has been achieved using external magnetic field along the  $a$ -axis, the comparison to the present results is still very instructive. It has been observed that the excitations of the  $ab$ -cycloid are dominated by a strong mode at  $2.25 \text{ meV}$  [31]. This frequency corresponds well to the excitation at  $21 \text{ cm}^{-1}$ , seen in Figs. 2,3.

In conclusion, we performed detailed polarization analysis of the electric and magnetic excitations in  $\text{TbMnO}_3$  in the high-field phase where spin cycloid rotates from  $bc$ - to  $ab$ -plane. The observed excitation at  $21 \text{ cm}^{-1}$  could not be described by purely magnetic contribution as was suggested previously. We argue that this excitation is the missing electro-active eigenmode of the spin cycloid. The weakness of this mode is in agreement with the Dzyaloshinskii-Moriya contribution to the dynamical magnetoelectric coupling in  $\text{TbMnO}_3$ , which represents a relativistic counterpart to the Heisenberg exchange mechanism.

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